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OXYGEN ISOTOPES IN CIRCUMSTELLAR Al_2O_3 GRAINS FROM METEORITES AND STELLAR NUCLEOSYNTHESIS

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ABSTRACT

We have found a ~ 3 μm Al_2O_3 grain (B39) in the Bishunpur LL3.1 chondrite that is enriched in ^{17}O by a factor of ~ 6.8 ($^{16}O/^{17}O = 385 \pm 9$) and depleted in ^{18}O by almost 60% ($^{16}O/^{18}O = 853 \pm 30$) relative to solar system oxygen and has an initial $^{26}Al/^{27}Al = 1.7 \pm 0.2 \times 10^3$, ~ 34 times greater than the initial solar system value. The isotopic compositions of B39 and two other Al_2O_3 grains previously reported from the Orgueil and Murchison meteorites show that these grains formed directly from the ejecta of low-mass AGB stars with C/O < 1. A simple theoretical analysis is presented showing that the oxygen systematics of the grains are a natural consequence of main-sequence evolution followed by first dredge-up. ^{26}Al is the result of third dredge-up. Circumstellar Al_2O_3 grains provide very precise isotopic data for stellar ejecta that complement spectroscopic observations of oxygen-rich stars. Isotopic differences indicate that the Al_2O_3 grains come from separate stars of different mass and initial oxygen composition that originated in molecular clouds different from the one in which the solar system formed.

Subject headings: circumstellar matter — dust, extinction — nuclear reactions, nucleosynthesis, abundances — stars: AGB and post-AGB — solar system: formation

1. INTRODUCTION

Until recently, all known types of presolar grains found in meteorites were carbon rich (diamond, silicon carbide, graphite, titanium carbide) and apparently formed in the ejecta of carbon stars (Anders & Zinner 1993; Ott 1993). However, astronomical observations show that much of the dust ejected from stars is oxygen rich (e.g., Gehrz 1989). Thus, meteorites should also contain presolar oxides. Finding presolar oxides has been difficult because the techniques used to separate carbon-rich grains from oxygen-rich minerals in stony meteorites destroy most oxides (Tang & Anders 1988; Amari, Lewis, & Anders 1994). Some refractory oxides survive these procedures, which has allowed the discovery of two presolar Al_2O_3 grains in the Orgueil (CI) and Murchison (CM2) meteorites (Huss, Hutcheon, & Wasserburg 1992; Hutcheon et al. 1994; Nittler et al. 1993). These grains have $^{16}O/^{17}O$ ratios a factor of 2 lower than solar [$(^{16}O/^{17}O)_\odot = 2610$], $^{16}O/^{18}O$ ratios equal to or slightly higher than solar [$(^{16}O/^{18}O)_\odot = 498$], and inferred $^{26}Al/^{27}Al$ initial ratios of $\sim 9 \times 10^{-4}$, 18 times the canonical solar system value of 5×10^{-5}. Their isotopic characteristics were interpreted as signatures of hydrogen burning via the CNO cycle (Hutcheon et al. 1994). We report on a third presolar Al_2O_3 grain (B39) from the Bishunpur (LL3.1) meteorite. The isotopic compositions of three presolar Al_2O_3 grains are compared with calculations of stellar nucleosynthesis and potential stellar sources are identified. We also discuss the implications of these grains for solar system and Galactic oxygen. A companion *Letter* (Boothroyd, Sackmann, & Wasserburg 1994) describes computational modeling of oxygen compositions in the envelopes of low- and intermediate-mass stars that can be used to identify possible sources of these grains.

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2. EXPERIMENTAL PROCEDURES

A sample of the Bishunpur meteorite was dissolved with a series of acid treatments (Huss & Lewis 1994) to produce a residue composed mainly of spinel ($MgAl_2O_4$), silicon carbide (SiC), hibonite ($CaAl_{12}O_{19}$), corundum (Al_2O_3), and TiO_2. Aliquots of this residue were deposited on cleaned gold foils. A JEOL 35F scanning electron microscope with an EDX X-ray detector was used to determine major and minor element abundances for each mineral grain (detection limits are $\sim 0.5\%$). The PANURGE IMS-3f ion microprobe was used to estimate trace-element abundances and to determine the isotopic compositions of oxygen and magnesium following procedures described in Hutcheon et al. (1994).

3. RESULTS

Oxygen isotopic compositions of 13 spinel, 14 hibonite, one TiO_2, and 27 Al_2O_3 grains were determined. Fifty-four grains had oxygen compositions within the range previously exhibited by minerals believed to have formed in the solar system (Fig. 1). Most of these grains are ^{16}O-enriched (up to 10%) relative to terrestrial oxygen and fall along the ^{16}O-mixing line defined by calcium-aluminum-rich inclusions (CAI) from carbonaceous chondrites (Clayton et al. 1977) and will not be discussed further here. One grain (B39) exhibited highly anomalous oxygen isotopes with $^{16}O/^{17}O = 385 \pm 9$ [$0.15 \times (^{16}O/^{17}O)_\odot$] and $^{16}O/^{18}O = 853 \pm 30$ [$1.7 \times (^{16}O/^{18}O)_\odot$]. Its magnesium isotope composition was $^{25}Mg/^{24}Mg = 0.1267 \pm 0.0023$; $^{26}Mg/^{24}Mg = 0.3728 \pm 0.0104$ (solar ratios are 0.12663 and 0.13932, respectively). The excess ^{26}Mg and an $^{27}Al/^{24}Mg$ ratio of 136 ± 14 give an initial $^{26}Al/^{27}Al$ ratio of $1.7 \pm 0.2 \times 10^{-3}$, ~ 34 times the canonical solar system value of 5×10^{-5}. The extreme oxygen isotopic composition and the high $^{26}Al/^{27}Al$ point to a formation site directly associated with stellar nucleosynthesis. Thus, B39 is most plausibly a circumstellar condensate.

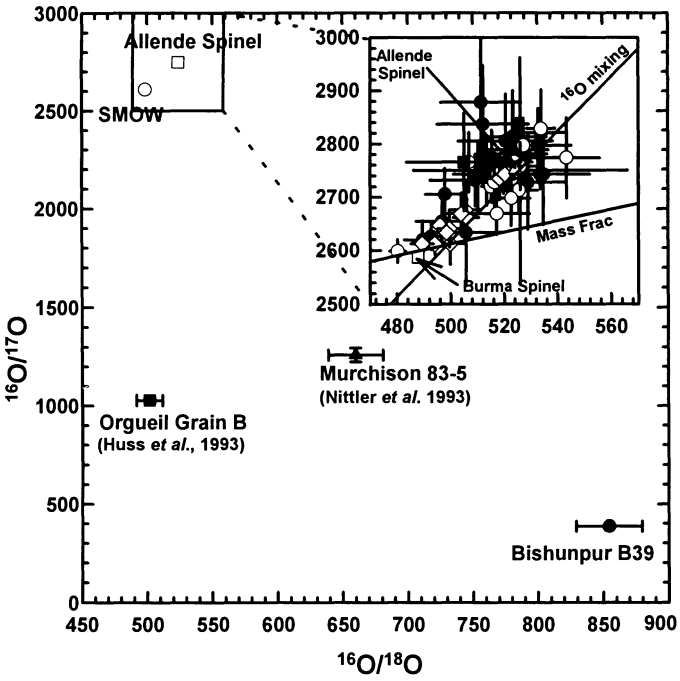

FIG. 1.—Oxygen isotope data for 55 oxide-mineral grains from the Bishunpur meteorite. Most data fall within the small box in the upper left (see inset) and lie along the ^{16}O-mixing line characteristic of meteoritic material formed in the solar system. Reference compositions for Allende meteoritic spinel, Burma terrestrial spinel, and standard mean ocean water (SMOW) are indicated. One Al_2O_3 grain, Bishunpur B39, is clearly distinct from the others. This grain and two others discovered previously are presolar oxide grains.

B39 is inferred to be an Al_2O_3 grain based on its X-ray spectrum, which showed only an aluminum peak, and the fact that oxygen was the only other major element present as determined from ion probe analysis. Approximate trace element abundances are 0.5% Mg, $\leq$0.05% Na, 0.03% Ca, 0.1% Ti, 0.004% V, 0.08% Cr, and 0.04% Fe by weight (no Si was detected). B39 is $\sim$3 μm in diameter, and its surface is similar to those of individual circumstellar SiC grains, which have been characterized as "platy" (Stone et al. 1991). The inferred abundance of presolar Al_2O_3 grains $>$1 μm is about 1% of the abundance of circumstellar SiC of the same size, or $\sim$5–10 ppb.

4. DISCUSSION

The oxygen isotopic ratios of grain B39 are more extreme than those of the previously recognized presolar Al_2O_3 grains, Orgueil B (Hutcheon et al. 1994) and Murchison 83-5 (Nittler et al. 1993), but all three grains show the same general features (Fig. 1): ^{17}O is enriched by factors of 2 to almost 7 relative to the solar composition, while ^{18}O is solar or slightly depleted. Similar trends are observed spectroscopically in the envelopes of red giant and oxygen-rich AGB stars (Harris & Lambert 1984; Harris, Lambert, & Smith 1988; Smith & Lambert, 1990) and in carbon-rich circumstellar shells (Kahane et al. 1992) and are consistent with theoretical expectations for such stars. The precisely determined oxygen isotope ratios and initial ^{26}Al/^{27}Al ratios of the Al_2O_3 grains permit us to examine in some detail the nucleosynthesis occurring in stars during the main sequence and later phases and to constrain the number and types of stars that might have produced the observed grains.

4.1. *Oxygen-Isotope Nucleosynthesis*

We focus here on production of ^{17}O and destruction of ^{18}O during main-sequence evolution and do not discuss production or destruction of oxygen isotopes in other stellar environments. During the main sequence, partial hydrogen burning via the CN cycle occurs in the stratified radiative region just outside of the convective core (Fig. 2). While the ^{16}O abundance outside the core remains essentially unchanged, ^{17}O is produced along with ^{13}C and ^{14}N, while ^{12}C, ^{15}N, and ^{18}O are destroyed (Rolfs & Rodney 1988; Dearborn 1992; El Eid 1994). This material is mixed to the surface in the red giant phase during the "first dredge-up," when the convective envelope extends downward to homogenize the outer $\sim$70%–80% of the star (Iben 1977; Dearborn 1992; El Eid 1994; Boothroyd et al. 1994).

At temperatures found at the base of the radiative zone (12–18 $\times$ 10^6 K), ^{17}O is produced via ^{16}O(p, γ)^{17}O and is destroyed via ^{17}O(p, γ)^{18}F(e^+ ν)^{18}O and ^{17}O(p, α)^{14}N. These three reaction rates are proportional to $\sim T^{\omega}$ ($\omega \approx 22$) and are comparable [assuming the ^{17}O(p, α)^{14}N rate given by Caughlan & Fowler 1988 with $f = 1$ or Landré et al. 1990 with $f_1 = 0.2$]. Since $^{16}X_0$ is $\sim$2600 $\times$ $^{17}X_0$, production of ^{17}O dominates destruction and large additions of ^{17}O are possible. The contribution of ^{17}O comes from a narrow zone near m_d (Fig. 2). In stars $\lesssim 3\ M_\odot$, the mean lifetimes ($\bar{\tau}_i$) for destruction of ^{16}O and ^{17}O are significantly longer than the time that the star is on the main sequence; thus their initial abundances are essentially unchanged. Under these conditions, the mass of ^{17}O in the convective envelope (E) after first dredge-up can be approximated by

$$^{17}M_E \approx {}^{17}X_0(m_t - m_d) + (17/16)^{16}X_0[t/(\bar{\tau}_{16}\,\omega\alpha)] \quad (1a)$$

with the number ratio of ^{17}O to ^{16}O as:

$$(^{17}O/^{16}O)_E \approx (^{17}O/^{16}O)_0 + t/[\bar{\tau}_{16}\,\omega(m_t - m_d)\alpha] \quad (1b)$$

where $\bar{\tau}_{16}$ is evaluated at the effective temperature and density at the dredge-up depth (T_d, ρ_d), t is the effective timescale for nucleosynthesis, $^iX_0$ is the initial mass fraction of "i," m_t and m_d are the mass of the star and the mass cut for first dredge-up, and $\alpha \equiv -(\partial \ln T/\partial m)_d$, which is of order unity. T_d increases

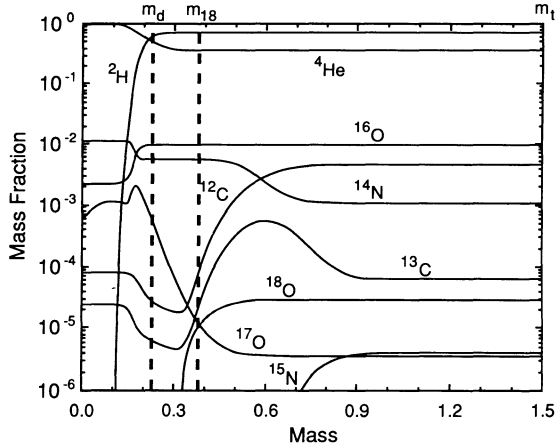

FIG. 2.—Abundance profiles are shown for a 1.5 $M_\odot$ star near the end of its main-sequence life (after Dearborn 1992). The convective core extends out to about 0.15 $M_\odot$. m_{18} is the depth at which ^{18}O begins to be significantly destroyed. The first dredge-up homogenizes the envelope down to mass m_d, producing higher ^{17}O and ^{13}C and lower ^{12}C and ^{18}O abundances than the initial values.

with stellar mass, and hence $\bar{\tau}_{16}$ rapidly decreases. This approximation fails for stars $\gtrsim 3\ M_\odot$, because (1) the ^{17}O destruction rate becomes 10–100 times greater than production at higher temperatures; (2) the convective envelope begins to extend into regions processed by the hotter, contracting core during the early main sequence; and (3) $t/\bar{\tau}_i$ approaches or exceeds unity. Approximately correct results for stars up to $\sim 3\ M_\odot$ can be obtained with parameters taken from stellar models in the literature. For example, for the $3\ M_\odot$ star of Iben (1965), taking the lifetime of the star (3.2×10^8 yr) for t, $T_d = 19 \times 10^6$ K and $\rho_d = 15$ g cm^{-3} (which gives $\bar{\tau}_{16} = 1.6 \times 10^9$ yr; Caughlan & Fowler 1988), $m_d = 0.54\ M_\odot$, and $\alpha \sim 1$, the calculated $^{16}O/^{17}O$ is ~ 230. This is within 50% of the result obtained in the detailed model of Boothroyd et al. (1994).

The (p, α) and (p, γ) reactions destroy ^{18}O 1000 times more efficiently than ^{16}O and ^{17}O at the temperatures of interest (Caughlan & Fowler 1988). The mass cut (m_{18}) below which ^{18}O is burned is well above the dredge-up depth (Fig. 2) as determined by $^{18}X \approx {}^{18}X_0\, e^{-t/\bar{\tau}_{18}}$ ($\bar{\tau}_{18}$ is obtained from the temperature and density structure of the star). The mass of ^{18}O in the envelope after first dredge-up is

$$^{18}M_E = (m_t - m_{18})^{18}X_0 \qquad (2a)$$

with the number ratio of ^{18}O to ^{16}O given by:

$$(^{18}O/^{16}O)_E = (m_t - m_{18})/(m_t - m_d)(^{18}O/^{16}O)_0\ . \qquad (2b)$$

Since the dilution factor $(m_t - m_{18})/(m_t - m_d)$ ranges from 0.6 to 0.9 for stars from 1 to $\sim 15\ M_\odot$, the ^{18}O abundance in the envelope after dredge-up will be $\sim 40\%$ to 10% lower than the initial abundance, with little dependence on the mass of the star (Dearborn 1992; Boothroyd et al. 1994).

The oxygen-isotope compositions of the three Al_2O_3 grains are compared with calculated compositions for envelopes of 1.2–3 $M_\odot$ stars after first dredge-up in Figure 3. Parameters for equations (1b) and (2b) were taken from the stellar evolution models of Boothroyd et al. (1994). The calculated compositions

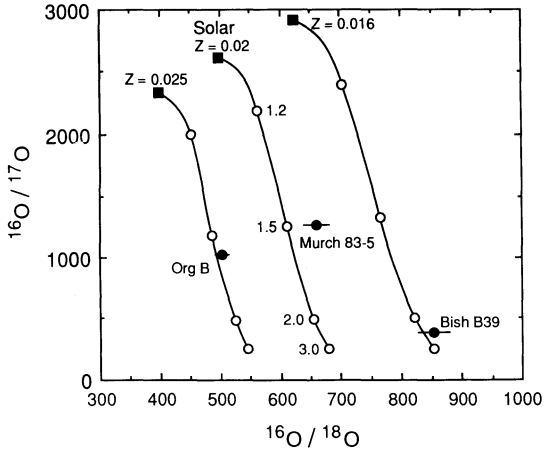

FIG. 3.—Oxygen isotopic compositions expected in the envelopes of 1–3 $M_\odot$ red giant stars after first dredge-up are compared to the compositions of three presolar Al_2O_3 grains. The family of curves shown were calculated from eqs. (1b) and (2b) with model parameters from Boothroyd et al. (1994). Each schematic curve was calculated for a different initial composition scaled with metallicity (Z) following Timmes et al. (1993). There is a strong dependence of $^{16}O/^{17}O$ on stellar mass, but only a weak dependence of $^{16}O/^{18}O$. Note that none of the curves passes through more than one of the measured compositions. Thus, the three Al_2O_3 grains must come from three distinct stars, two of roughly 1.5 $M_\odot$ and one somewhat more massive.

(Fig. 3) are similar to the exact results obtained from detailed calculations (cf. Fig. 4 of Boothroyd et al. 1994). Stellar nucleosynthesis during the main sequence can lower the $^{16}O/^{17}O$ ratio in the envelope by factors up to ~ 20, but the $^{16}O/^{18}O$ ratio changes by less than 50%. The $^{16}O/^{17}O$ ratio decreases rapidly with increasing stellar mass up to $\sim 3\ M_\odot$, but increases again with further increases in mass (Dearborn 1992; El Eid 1994; Boothroyd et al. 1994). The $^{16}O/^{17}O$ ratios indicate that two Al_2O_3 grains come from stars of $\sim 1.5\ M_\odot$, while B39 comes from a star of ~ 2 or 4–7 $M_\odot$ (see also Boothroyd et al. 1994). The second and third dredge-ups have little effect on the oxygen isotopic composition of the envelope. This is confirmed by observations of MS and S stars that have experienced third dredge-up (Smith & Lambert 1990) and the envelopes of carbon stars (Kahane et al. 1992) and by calculations of Boothroyd et al. (1994).

The Al_2O_3 grains exhibit a range in $^{16}O/^{18}O$ that is larger than predicted for a suite of stars with the same starting composition (Fig. 3). The predicted range in $^{16}O/^{18}O$ does not increase significantly for stars up to $\sim 15\ M_\odot$ (Dearborn 1992; El Eid 1994; Boothroyd et al. 1994). Thus, a different starting composition is required to produce each of the three Al_2O_3 grains, which in turn implies a different stellar source for each grain. Further, the three parent stars apparently were not related to one another or, given the relative time scales of stellar evolution, to the molecular cloud from which the Sun formed.

4.2. *Synthesis of* ^{26}Al *and Grain Formation*

The Al_2O_3 grains must have formed in a circumstellar environment with sufficient density to permit grain formation, with C/O < 1, and with ^{26}Al present ($^{26}Al/^{27}Al \approx 10^{-3}$). ^{26}Al is produced via $^{25}Mg(p, \gamma)^{26}Al$ at temperatures $\gtrsim 30 \times 10^6$ K in the hydrogen shell (cf. Forestini, Paulus, & Arnould 1991). During the thermally pulsing AGB phase, convective mixing driven by He shell flashes (the third dredge-up) brings ^{12}C from the helium shell and ^{26}Al from the hydrogen shell into the envelope, increasing C/O to eventually produce a carbon star and giving an $^{26}Al/^{27}Al$ ratio of up to 10^{-2}. The corresponding shift in $^{25}Mg/^{24}Mg$ to be expected from this process is

$$(^{25}Mg/^{24}Mg)/(^{25}Mg/^{24}Mg)_0 = 1 - (^{26}Al/^{27}Al)(^{27}Al/^{25}Mg)_0\ . \qquad (3)$$

Early pulses, where the envelope has C/O < 1, give $^{26}Al/^{27}Al$ ratios of 10^{-4} to 10^{-3} (Wasserburg et al. 1994), consistent with inferred ratios in the Al_2O_3 grains and no measurable deficit of ^{25}Mg in grain B39. We cannot determine whether the ^{26}Al was alive or extinct in the grains when they were added to the protosolar cloud.

Mass-loss rates during and after first dredge-up are several orders of magnitude below those where grain formation is considered possible. However, shortly before becoming carbon stars, low-mass AGB stars may become Mira variables, which have observed mass loss rates of up to $10^{-6}\ M_\odot$ yr^{-1} and infrared excesses indicating grain formation (Willems & de Jong 1988). It is reasonable to suppose that the Al_2O_3 grains formed during the early AGB phase when there is substantial mass loss, after several He shell flashes supplied some ^{26}Al to the envelope, but prior to the time when the AGB star becomes a carbon star with C/O > 1 (Busso et al. 1994). Theoretical models suggest that mass-loss rates during the late AGB, with C/O > 1, might reach $10^{-4}\ M_\odot$ yr^{-1} (Bowen & Willson 1991)

which may explain why presolar SiC is a factor of 10^2 more abundant than Al_2O_3.

4.3. *Contributions to the Solar System*

The presence of short-lived nuclei (^{26}Al, ^{107}Pd, ^{60}Fe) in the early solar system indicates that freshly synthesized material from stellar sources was added to the molecular cloud from which the solar system formed. AGB star(s) in the neighborhood of the molecular cloud have been proposed as a source for these short-lived nuclei (Cameron 1993; Wasserburg et al. 1994). A molecular cloud is likely to encounter an AGB star some time during its 10^8 yr lifetime (Kastner & Myers 1994). The discovery in meteorites of ^{17}O-rich, ^{18}O-depleted Al_2O_3 grains with ^{26}Al excesses demonstrates that AGB stars are capable of producing and supplying oxide grains to the protosolar mix. However, the observed abundance of presolar Al_2O_3 grains is much too low for them to be the carriers of the ^{26}Al inferred to have been present in the early solar system (Hutcheon et al. 1994).

Most solar system material has been extensively processed and no longer contains primitive presolar grains. However, refractory calcium-aluminum inclusions (CAI) are enriched in ^{16}O by up to $\sim 5\%$. These excesses have long been interpreted in terms of a presolar ^{16}O-rich carrier (Clayton et al. 1977). Although CAI are also anomalous in many other elements, the anomaly patterns do not point to specific stellar sources. The absence of specific stellar isotopic signatures, the rather small isotopic anomalies, and the igneous textures and large size of the CAI (millimeters to centimeters), indicate that these objects formed in the solar system. In light of the indications that an ^{16}O carrier was present in the early solar system, it is surprising that the first recognized presolar oxide grains are ^{17}O rich. The presence of identifiable ^{16}O-rich carrier grains has not been established. It is possible that the ^{16}O enrichments observed in meteorites are not of nucleosynthetic origin but arose from non–mass-dependent isotope fractionation (Thiemens & Heidenreich 1983).

There is a discrepancy between the $^{18}O/^{17}O$ ratio of the solar system and ratios now observed in molecular clouds, both at the Galactic center and throughout the Galaxy. The $^{18}O/^{17}O$ from diverse molecular clouds is constant at 3.65 ± 0.15 (Penzias 1981; Wannier 1989), a factor of ~ 1.5 below the solar system value. If the oxygen isotopic composition of the gas properly represents the whole molecular cloud, then we are left with a conundrum concerning whether the $^{18}O/^{17}O$ for the solar system reflects the isotopic composition at about $\frac{2}{3}$ the Galactic age or whether there is some "special" component of oxygen in the solar system that makes it distinct.

5. SUMMARY AND CONCLUSIONS

A presolar Al_2O_3 grain from the Bishunpur meteorite was found with a large excess of ^{17}O and a deficit of ^{18}O relative to solar abundances. These characteristics are similar to those of two previously recognized presolar Al_2O_3 grains and point to red giant and AGB stars as sources. Consideration of oxygen synthesis in stars shows that the three Al_2O_3 grains must have come from separate, unrelated stars. The presence of ^{26}Al in the grains and their oxygen-rich chemistries point to origins in oxygen-rich AGB stars.

After submission of this *Letter*, an abstract by Nittler et al. (1994) reported the discovery of additional oxide grains with oxygen isotopic anomalies. Many of these grains had oxygen isotopic compositions similar to those discussed here and may be explained by an AGB source. However, some were distinctly different and require other nucleosynthetic mechanisms.

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